Non-Locality and Strong Coupling in the Heavy Fermion Superconductor $CeCoIn_5$: A Penetration Depth Study

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(Dated: February 1, 2008)

We report measurements of the magnetic penetration depth λ in single crystals of $CeCoIn_5$ down to ∼0.14 K using a tunnel-diode based, self-inductive technique at 28 MHz. While the in-plane penetration depth tends to follow a power law, $\lambda_{//} \sim T^{3/2}$, the data are better described as a crossover between linear ($T \gg T^*$) and quadratic ($T \ll T^*$) behavior, with T^* the crossover temperature in the strong-coupling limit. The *c*-axis penetration depth $\lambda_\perp$ is linear in T, providing evidence that $CeCoIn_5$ is a *d*-wave superconductor with line nodes along the *c*-axis. The different temperature dependences of $\lambda_{//}$ and $\lambda_\perp$ rule out impurity effects as the source of T^*.

The compounds $CeMIn_5$ (M = Co, Ir, Rh) have recently been added to the heavy-fermion family, and have attracted much interest due to their similarity with the cuprates: quasi-2D structure and proximity to magnetic order [1]. $CeCoIn_5$, in particular, is a good candidate for study: its superconductivity is not sensitive to small changes in unit-cell volume or composition, unlike $CeCu_2Si_2$, and it has the highest T_c (∼2.3 K) among the heavy-fermion superconductors. $CeCoIn_5$ has tetragonal $HoCoGa_5$ crystal structure, consisting of alternating layers of $CeIn_3$ and '$CoIn_2$' [1]. De Haas-van Alphen (dHvA) data revealed that the Fermi surface (FS) is quasi-2D, with an open 2D undulating cylinder extending along the [001] direction, as well as the large effective masses of electrons on this FS [2].

Recently, there has been mounting evidence for unconventional superconductivity in $CeMIn_5$. Specific heat data reveal a T^2 term at low temperature, consistent with the presence of line nodes in the superconducting energy gap [3]. Thermal conductivity measurements with in-plane applied field show four-fold symmetry, consistent with nodes along the ($\pm\pi$, $\pm\pi$) positions [4]. NQR measurements show that there is no Hebel-Slichter peak just below T_c [5]. Below T_c the spin susceptibility is suppressed, indicating singlet pairing [5, 6]. However, there are some ambiguities in some of the measurements. Thermal conductivity data yield a $T^{3.37}$ low-temperature behavior, that the authors claim is close to T^3 behavior predicted for unconventional superconductors with line nodes in the clean limit [3]. NQR measurements did not show the T^3 low-temperature behavior of $1/T_1$ that is expected for a line node gap; instead $1/T_1$ saturates below 0.3 K [5]. Microwave measurements down to ∼0.2 K showed a non-exponential behavior, and the authors claimed that $\lambda(T) \sim T$ below 0.8 K [7], though the data clearly show some curvature in that temperature range. Further, the field was applied along the *ab*-plane, so the shielding currents have both in-plane and inter-plane components. In this paper, we present high-precision measurements of in-plane $\lambda_{//}$ and inter-plane $\lambda_\perp$ penetration depths of $CeCoIn_5$ at temperatures down to 0.14 K. We find that $\lambda_{//}$ is best treated as a crossover from $\sim T$ to $\sim T^2$ at a temperature T^*. Combined with the result that $\lambda_\perp \propto T$, this gives strong evidence for non-local behavior in a *d*-wave superconductor as predicted by Kostzin and Leggett [8].

Details of sample growth and characterization are described in Refs. [1, 9]. Measurements were performed utilizing a 28 MHz tunnel diode oscillator [10] with a noise level of 1 part in 10^9 and low drift. The magnitude of the ac field was estimated to be less than 5 mOe. The cryostat was surrounded by a bilayer Mumetal shield that reduced the dc field to less than 1 mOe. The sample was aligned inside the probing coil in two directions: (1) *ab* plane perpendicular to the rf field, measuring the in-plane penetration depth $\lambda_{//}$ (screening currents in the *ab* plane); or (2) with the rf field parallel to the plane, giving a combination of $\lambda_{//}$ and $\lambda_\perp$. The sample was mounted, using a small amount of GE varnish, on a rod made of nine thin 99.999% Ag wires embedded in Stycast 1266 epoxy. The other end of the rod was thermally connected to the mixing chamber of an Oxford Kelvinox 25 dilution refrigerator. The sample temperature is monitored using a calibrated RuO_2 resistor at low temperatures (T_{base} - 1.8 K), and a calibrated Cernox thermometer at higher temperatures (1.3 K - 2.5 K). We report data only for

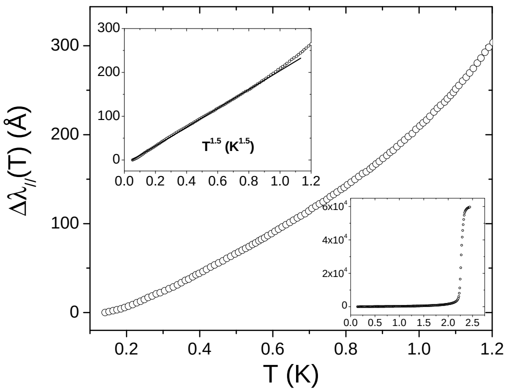

FIG. 1: Low-temperature dependence of the in-plane penetration depth $\Delta\lambda_{//}(T)$. Lower inset shows $\Delta\lambda_{//}(T)$ over the full temperature range. Upper inset shows $\Delta\lambda_{//}(T)$ vs $T^{1.5}$ in the temperature range (0.14-1.13) K. The solid line is a guide to the eye.

$T \geq 0.14$ K. The value of T_c was determined from magnetization measurements to be 2.3 K, identical to the previously reported value [3].

The deviation $\Delta\lambda(T) = \lambda(T) - \lambda(0.14\text{ K})$ is proportional to the change in resonant frequency $\Delta f(T)$, with the proportionality factor G dependent on sample and coil geometries. For a square sample of side $2w$, thickness $2d$, demagnetization factor N, and volume V, G is known to vary as $G \propto R_{3D}(1\text{-}N)/V$, where $R_{3D} = w/[2(1+(1+2d/w)^2)\arctan(w/2d)\text{-}2d/w]$ is the effective sample dimension [11]. For our sample $2w \approx 0.73$ mm and $2d \approx 0.09$ mm. We determine G from a single-crystal sample of pure Al by fitting the Al data to extreme non-local expressions and then adjusting for relative sample dimensions. Testing this approach on a single crystal of Pb, we found good agreement with conventional BCS expressions.

Fig. 1 shows $\Delta\lambda_{//}(T)$ as a function of temperature. We see that $\Delta\lambda_{//}(T)$ varies strongly at low temperatures, inconsistent with the exponential behavior expected for isotropic s-wave superconductors. On the other hand, the variation is not linear, but has an obvious upward curvature, unlike the low-temperature behavior expected for pure d-wave superconductors. A fit of the low temperature data to a variable power law, $\Delta\lambda_{//}(\text{T}) = a + bT^n$ yields $n = 1.43 \pm 0.01$ for sample 1 and 1.57 ± 0.01 for sample 2. The upper inset of Fig. 1 shows this approximate $T^{3/2}$ behavior for sample 1. Kosztin *et. al.* [12, 13, 14] have proposed a theory that gives a $T^{3/2}$ term from the gradual evolution of the pseudogap above T_c to the superconducting gap below T_c. While resistivity measurements suggest the possibility of a pseudogap in $CeCoIn_5$ [15], which renders this interpretation feasible, a decrease in Knight shift was observed only starting at T_c [6]. We take the latter to rule out a pseudogap mechanism.

Before considering novel excitation processes, we note the important distinction between $\Delta\lambda(T)$, which is directly measured, and the superfluid density $[\rho(T) = \lambda^2(0)/\lambda^2(T)]$ which can be inferred only with the knowledge of $\lambda(0)$ [16]. In the d-wave model, even if ρ varies strictly with T, i.e. ρ=1-$\alpha T/T_c$, the penetration depth is non-linear: $\lambda(T) = \lambda(0)[1+1/2\ (\alpha T/T_c) + 3/8\ (\alpha T/T_c)^2 + ...]$. Hence there is always a quadratic component to λ whose strength depends on α, which in the d-wave model, is inversely proportional to $d\Delta(\theta)/d\theta|_{node}$, the angular slope of the energy gap at the nodes [17]. If $\rho(T)$ is linear in T, there is no need to invoke another mechanism.

To extract the in-plane superfluid density from our data, we need to know $\lambda_{//}(0)$. For a quasi-2D superconductor with a cylindrical Fermi surface and the material parameters in Ref. 3 [18], we obtain $\lambda_{//}(0) = 2600$ Å, considerably larger than the experimentally obtained value of 1900 Å [7]. This along with a large heat-capacity jump at T_c leads us to consider strong-coupling corrections as listed below [19, 20]:

$$\eta_{Cv}(\omega_0) = 1 + 1.8(\frac{\pi T_c}{\omega_0})^2(\ln(\frac{\omega_0}{T_c}) + 0.5); \qquad (1)$$

$$\eta_{\Delta}(\omega_0) = 1 + 5.3(\frac{T_c}{\omega_0})^2 \ln(\frac{\omega_0}{T_c}); \qquad (2)$$

$$\eta_{\lambda}(\omega_0) = \frac{\sqrt{1 + (\frac{\pi T_c}{\omega_0})^2(0.6\ln(\frac{\omega_0}{T_c}) - 0.26)}}{1 + (\frac{\pi T_c}{\omega_0})^2(1.1\ln(\frac{\omega_0}{T_c}) + 0.14)}; \qquad (3)$$

each η represents the correction to the corresponding BCS value. If we take the experimental value of $\Delta C/\gamma T_c$ = 4.5 [1], then Eq. 1 gives the characteristic (equivalent Einstein) frequency $\omega_0 = 9.1$ K and $\lambda^{sc}_{//}(0) = 1500$ Å. However, Petrovic *et. al.* [1] argued that since C/T increases with decreasing temperature, the specific heat coefficient γ is temperature-dependent below T_c. This effect calls into question simple estimates of strong-coupling corrections for $CeCoIn_5$. A better estimate is to use $\Delta C/\Delta S$, where ΔS is the measured change in entropy of the sample from $T = 0$ to T_c. Ref. 1 then gives $\Delta C/\Delta S = 2.5$, so that $\omega_0 = 17.9$ K, resulting in $\Delta^{sc}_0 = 2.1 k_B T_c$ and $\lambda^{sc}_{//}(0) = 2000$ Å. On the other hand, the larger ΔC of Ref. 21 yields $\Delta C/\Delta S = 2.8$ and $\omega_0 = 15.4$ K, leading to $\Delta^{sc}_0 = 2.2 k_B T_c$ and $\lambda^{sc}_{//}(0) = 1900$ Å. These values of $\lambda^{sc}_{//}(0)$ are close to that obtained by Ormeno *et. al.* [7].

Although we will argue that non-local effects are important, we will refer to $(\lambda_{//}(0)/\lambda_{//}(T))^2$ as the "superfluid density." Fig. 2 shows the calculated behavior

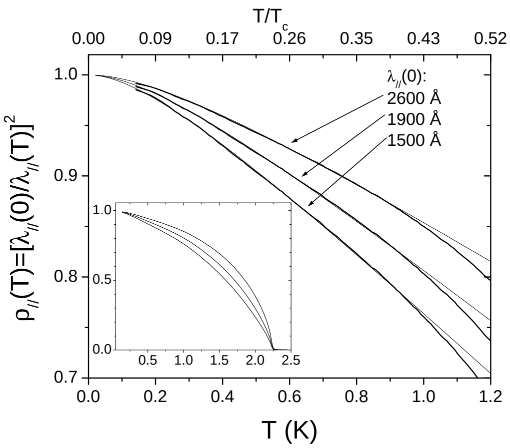

FIG. 2: Low-temperature in-plane superfluid density $\rho_{//}$(T) = $[\lambda^2_{//}(0)/\lambda^2_{//}(\mathrm{T})]$ calculated from $\Delta\lambda_{//}(T)$ data in Fig. 1 (thick lines). The thin lines correspond to fits to data using Eq. 5, using three values of $\lambda_{//}(0)$. Inset shows $\rho_{//}$(T) over the full temperature range.

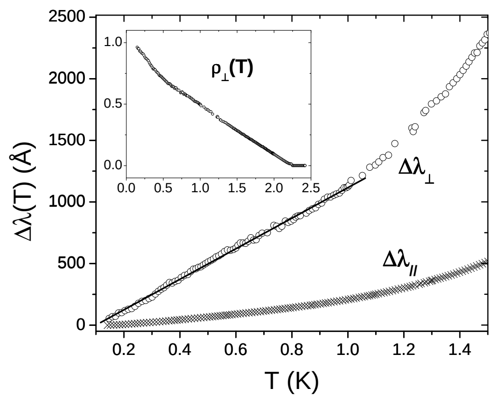

FIG. 3: Low-temperature dependence of inter-plane (open circles) penetration depth $\Delta\lambda_\perp(T)$, after subtracting the in-plane component. In-plane $\Delta\lambda_{//}(T)$ (crosses) data is also shown for comparison. Solid line is a linear fit from 0.14 K to 1 K. Inset shows inter-plane superfluid density $\rho_\perp(T)$ for the whole temperature range.

of that quantity using the three values of $\lambda_{//}(0)$ obtained above. We follow the procedure in Ref. 16 to compute the experimental superfluid density, using the $T^{3/2}$ fit to estimate the small difference between $\lambda_{//}(0)$ and $\lambda_{//}$(0.14 K). In each case, $\rho(T)$ is clearly not linear in T.

Non-linearity in $\rho(T)$ can arise from a crossover from an intermediate-temperature (pure) linear-T behavior to, for example, low-temperature (impurity-dominated) quadratic behavior as pointed out by Hirschfeld and Goldenfeld [22]. They interpolated between these two regions using

$$\lambda = \lambda_0 + bT^2/(T^* + T), \tag{4}$$

where T^* is the crossover temperature. In terms of superfluid density, one obtains [16]

$$\Delta\rho_{//}(T) = \frac{\alpha T^2/T_c}{T^* + T}, \tag{5}$$

where T^* depends on impurity concentration.

A much more provocative source of the crossover of Eq. 5 was suggested by Kosztin and Leggett [KL] [8], who showed that for d-wave superconductors, nonlocal effects change the linear behavior to quadratic below a crossover temperature $T^*_{nonlocal} = \Delta_0\xi_{//}(0)/\lambda_{//}(0)$.

The solid lines in Fig. 2 are fits to Eq. 5 and are very good for all three values of $\lambda_{//}(0)$. The value of α varies from ∼0.5 to 0.7, the smallest value of α belonging to the largest value of $\lambda_{//}(0)$. The value of α obtained is similar to that found for $YBa_2Cu_3O_{6.95}$ ($\alpha \sim 0.6$) [23, 24], but smaller than that of $Tl_2Ba_2CuO_{6+\delta}$ ($\alpha \sim 1.0$) [25] and K-$(ET)_2Cu[N(CN)_2]Br$ ($\alpha \sim 1.2$) [16]. The value of T^* varies less across the three $\lambda_{//}(0)$ values, from 0.32 K to 0.42 K. These values of T^*/T_c (∼0.14 - 0.18) differ from the cuprates [23, 25, 26] and the organic superconductor K-$(ET)_2Cu[N(CN)_2]Br$ (∼0.05), where impurity scattering is presumed to be the source. Further, Ref. 3 puts an upper limit of 20 ppm on the impurity concentration. In the dirty d-wave model [22], this gives the unitary-limit scattering rate $\Gamma \sim 1.5 \times 10^8$ s^{-1}, which yields an upper limit for $T^* \sim 65$ mK. This is about 5 times smaller than the experimentally obtained values above, suggesting that the sample is too clean for the dirty d-wave model to be applicable.

Having ruled out impurity scattering, we turn to nonlocal electrodynamics as the source of the crossover in $\rho_{//}(T)$. For a d-wave superconductor *with line nodes along the c-axis*, nonlocality is expected to be relevant only when the applied magnetic field is oriented parallel to the c-axis, while the effect of impurities should not depend on the orientation of the field. As KL noted, if T^* is noticeably smaller for $H \perp c$ than for $H//c$ we may conclude that the observed effect is due mainly to nonlocal electrodynamics and not to impurities. For $H \perp c$, screening currents flow both parallel and perpendicular to the c-axis, mixing $\lambda_{//}$ and $\lambda_\perp$ with the frequency shift given by $\frac{\Delta f_\perp}{f_0} = \frac{V}{2V_0}(\frac{\lambda_{//}}{d} + \frac{\lambda_\perp}{w})$ [11], where V_0 is the effective coil volume and f_0 the resonant frequency with the sample absent. In order to extract $\lambda_\perp$ we subtract out the $\lambda_{//}$ component from $\Delta f_\perp$. Fig. 3 shows the inter-plane penetration depth $\lambda_\perp$ of $CeCoIn_5$ down to 0.14 K. It is clearly linear in T from 0.14 K to 1 K. To obtain the superfluid density, we estimate $\lambda_\perp(0)$ from the H_{c2} anisotropy of ∼2.3 [21], and the fact that $\lambda(0) \propto \sqrt{H_{c2}(0)}$ [27], obtaining $\lambda_\perp(0) \sim 2700$ Å. This is close to the value of ∼2800 Å obtained from microwave mea-

surements in the planar geometry [28]. If we fit $\lambda_\perp(T)$ to Eq. 4, we find $T^*_\perp \lesssim 0.15$ K, significantly smaller than 0.32 K obtained for the in-plane case. This satisfies the Kosztin-Leggett test and indicates that the superfluid response of $CeCoIn_5$ is governed by nonlocal electrodynamics. This is also strong evidence that *$CeCoIn_5$ is a d-wave superconductor with line nodes along the c-axis.* Sr_2RuO_4 failed this test [10] because its line nodes are horizontal instead of vertical. Kusunose and Sigrist argued that horizontal line nodes give power-law behaviors with less angular dependence for any inplane direction of the screening currents, and hence applied field [29]. A calculation of $\rho_\perp$ is shown in the inset of Fig. 3: the upturn below 0.5 K is an artifact of the choice of $\lambda_c(0)$. A larger value of $\lambda_c(0)$ would remove this feature, but there is no justification for doing so.

As a final test of the non-local scenario, we estimate $T^*_{//}$ using strong-coupling parameters. From the measured $H_{c2}(0)[001]$ value of 49.5 kOe, the coherence length $\xi_{//}(0)$ is calculated to be 82 Å [21]. Together with the earlier-derived values of $\Delta_0^{sc} = 2.2 k_B T_c$ and $\lambda^{sc}_{//}(0) = 1900$ Å, we find the strong-coupling nonlocal crossover temperature $T^*_{nonlocal} = \Delta_0^{sc}\ \xi_{//}(0)/\lambda^{sc}_{//}(0) = 0.22$ K. Using a weak-coupling d-wave $\Delta(0) = 2.14 k_B T_c$, we find $T^*_{nonlocal} = 0.26$ K. We regard either value to be satisfactorily close to the experimental value of 0.32 K. Note that the value of $\xi_{//}(0)$ is different from the calculated BCS value of 58 Å [3] or the strong-coupling corrected value of ~ 50 Å [19]. This is not surprising since the BCS expressions [19] assume a spherical FS, while LDA band structure reveals a very complicated FS with contributions from three different bands [30].

In conclusion, we report measurements of the magnetic penetration depth λ in single crystals of $CeCoIn_5$ down to $\sim$0.14 K using a tunnel-diode based, self-inductive technique at 28 MHz. The in-plane penetration depth ($\lambda_{//}$) exhibits a crossover between linear (at high T) and quadratic (low T) behavior with a crossover temperature $T^*_{nonlocal} \approx 0.32$ K. Such behavior can arise in a superconductor with nodes in the gap either in a dirty d-wave model or from nonlocal electrodynamics. The linear low-temperature dependence of the c-axis penetration depth $\lambda_\perp$ strongly favors the nonlocal model with line nodes parallel to the c-axis. We also demonstrate that strong-coupling corrections are required to reconcile various experimentally determined superconducting parameters.

One of the authors (E.E.M.C.) acknowledges R. Prozorov and Q. Chen for useful discussions. This work was supported by the NSF through Grant No. DMR9972087. Work at Los Alamos was performed under the auspices of the U.S. Department of Energy.

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